REMOVAL OF CRUDE OIL FROM AQUEOUS SOLUTION USING LAURIC ACID MODIFIED OIL PALM LEAVES ADSORBENT

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For my beloved family,

Thanks for everything.

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ABSTRACT

Recently, the abundant and biodegradable biomass was reported to be convenient for oil spill clean–up due to its low-cost and environmental friendly. One of the agricultural wastes, namely oil palm leaves (OPL) were chemically modified by a fatty acid, lauric acid (LA) and employed as an adsorbent to remove crude oil from aqueous solution. The textural and surface properties of the lauric acid modified oil palm leaves (OPL_{LA}) were characterized by FTIR, FESEM, and N_2 adsorption. Thermal stability of the adsorbents was characterized by TGA. The effect of several parameters such as solution pH, loading of adsorbent, contact time, initial crude oil concentration, and adsorption temperature on crude oil adsorption were investigated. It was found that addition of LA created non-polar layer on oil palm leaves surface, thus endowing OPL_{LA} with much better adsorption capacity for crude oil removal from aqueous solution. The adsorption was found to be dependent on all the studied parameters. The kinetic study revealed that adsorption process was rapid and obeyed pseudo second-order model. The equilibrium time was short and could be achieved within 20 minutes. Isotherm study indicated that the oil adsorption was fitted well by Freundlich model. The maximum adsorption capacity of OPL_{LA} controlled by the solubilization mechanism was 1176.00 ± 12.92 mg g^{-1} with the removal percentage of 42.00 ± 0.46 % at 303 K. The potential of OPL_{LA} to adsorb crude oil from crude oil contaminated seawater achieved 34.02 ± 0.45 % removal with the maximum adsorption capacity of 952.66 ± 11.31 mg g^{-1}. These results demonstrated the potential of OPL_{LA} as an alternative low-cost, biodegradable and effective adsorbent for oil spill clean-up.

ABSTRAK

Kebelakangan ini, biomas yang banyak dan boleh terbiodegradasi telah dilaporkan berguna untuk pembersihan tumpahan minyak disebabkan oleh ianya yang berkos rendah dan mesra alam. Salah satu daripada bahan sisa pertanian iaitu daun kelapa sawit (OPL) telah diubahsuai secara kimia dengan menggunakan asid laurik (LA) dan dijadikan sebagai bahan penjerap untuk pengasingan minyak mentah daripada larutan akueus. Tekstur dan sifat permukaan daun kelapa sawit yang telah diubahsuai menggunakan asid laurik (OPL_{LA}) telah dicirikan dengan menggunakan FTIR, FESEM, dan penjerapan N_2. Kestabilan terma bahan-bahan penjerap juga telah dicirikan menggunakan TGA. Kesan beberapa parameter seperti pH larutan, muatan bahan penjerap, masa sentuh, kepekatan awal minyak mentah dan suhu penjerapan ke atas penjerapan minyak mentah telah dikaji. Didapati bahawa penambahan LA telah menyediakan lapisan tidak berkutub pada permukaan daun kelapa sawit lalu menghasilkan OPL_{LA} yang mempunyai kapasiti penjerapan yang lebih baik untuk pengasingan minyak mentah daripada larutan akueus. Hasil kajian mendapati proses penjerapan adalah dipengaruhi oleh semua parameter yang dikaji. Kajian kinetik menunjukkan bahawa proses penjerapan adalah cepat dan mematuhi model kinetik tertib-kedua-pseudo. Masa keseimbangan adalah singkat dan boleh dicapai dalam tempoh 20 minit. Kajian isoterma mempamerkan bahawa penjerapan minyak adalah menepati model Freundlich. Kapasiti penjerapan maksimum OPL_{LA} yang dikawal oleh mekanisme penjerapan-pelarutan adalah 1176.00 ± 12.92 mg g^{-1} dengan peratus penyingkiran sebanyak 42.00 ± 0.46 % pada 303 K. Potensi OPL_{LA} untuk menjerap minyak mentah daripada air laut yang dicemari minyak mentah telah mencapai peratus penyingkiran sebanyak 34.02 ± 0.45 % dengan kapasiti penjerapan maksimum sebanyak 952.66 ± 11.31 mg g^{-1}. Hasil kajian telah menunjukkan potensi OPL_{LA} sebagai alternatif kepada bahan penjerap yang murah, boleh terdegradasi dan efektif untuk pembersihan tumpuhan minyak.

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LIST OF ABBREVIATIONS

BAF	-	Biological aerated filter
BET	-	Brunauer Emmett Teller
BRHA	-	Black rice husk ash
CRH	-	Carbonized rice husks
CFU	-	Compact flotation unit
EFB	-	Empty fruit bunch
FESEM	-	Field emission scanning electron microscopy
FTIR	-	Fourier transform infrared spectroscopy
HA	-	Hydrophobic aerogel
HAS	-	Hydrophobic aquaphyte
HCP	-	Hydrogel of chitosan based on polyacrylamide
LA	-	Lauric acid
MF	-	Microfiltration
NF	-	Nanofiltration
OGSD	-	Oleic acid-grafted sawdust
OPL	-	Oil palm leaves
OPL_{LA}	-	Oil palm leaves modified with lauric acid
RO	-	Reverse osmosis
RWNM	-	Recycled wool-based nonwoven material
SBRs	-	Sequencing batch reactors
SDS	-	Sodium dedocyl sulphate
TGA	-	Thermagravimetric analysis
UF	-	Ultrafiltration
UV	-	Ultraviolet
WPG		Weight percent gain

LIST OF SYMBOLS

E_a	-	Activation energy
q_e	-	Adsorption capacity at equilibrium
q_t	-	Adsorption capacity at specific time
C_e	-	Concentration of adsorbate
R_L	-	Dimensionless constant separation factor
ΔH^o	-	Enthalpy change
ΔS^o	-	Entropy change
ΔG^o	-	Free Gibbs energy
R	-	Gas constant
g	-	Gram
k_2	-	Ho-pseudo-second-order constant
C_o	-	Initial Concentration
J	-	Joule
K	-	Kelvin
k	-	Kilo
k_1	-	Lagergren-pseudo-first-order constant
K_L	-	Langmuir adsorption constant
L	-	Litre
Mg	-	Magnesium
m	-	Mili
MnT	-	Million tonnes
min	-	Minute
B	-	Temkin constant
q_m	-	Theoritical maximum adsorption capacity

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CHAPTER 1

INTRODUCTION

1.1 Background of Study

Petroleum is an important energy source and raw chemical material. Nowadays, rapid growth in economy and civilization developement has led to the tremendous consumption of petroleum products. With approximately 10 million tons of petroleum products and derivatives being used daily worldwide (Abdullah *et al*., 2010), the world is facing a great risk of petroleum products contamination in the environment. Oil and chemical spills accidents can be caused by human mistakes and carelessness, deliberate acts such as vandalism, war and illegal dumping, or by natural disasters such as hurricanes or earthquakes. Offshore and shoreline waters can be polluted by oil drilling operations, accidents involving oil tankers, runoffs from offshore oil explorations, and spills from tanker loading and unloading operations (Vlaev *et al*., 2011). Inland water bodies can be polluted by leaking of oil through pipeline corruptions, runoff from oil fields and refinery areas and process effluent from petrochemical plants.

Massive marine oil spills have occured frequently and resulted in a great deal of damage to the marine, coastal and terrestrial habitats, economical impacts on fisheries, mariculture and tourism and loss of energy source. Between 1974 and 1994, there were 174 major oil spills worlwide (Fingas, 2000). The cost just to clean up the contaminated site may go as high as \$20-\$200 per litre in USA and Canada, depending on the location and type of oil spilled (Abdullah *et al*., 2010).

An extensive literature has been devoted to the removal of oil from aqueous solution. These studies almost exclusively use either inorganic mineral materials, organic synthetic product or organic natural materials as the adsorbent (Teas *et al*., 2001). However, the use of various types of natural organic adsorbents, such as wool (Radetic *et al*., 2008), lemon peel (Bhatnagar *et al*., 2010), cotton (Deschamps *et al*., 3002), rice husk (Kumagai *et al*., 2007), and baggase (Sun *et al*., 2004) are particularly interesting because of their relatively high adsorption capacities, biodegradability and cost effectiveness compared to the synthetic polymeric fibers that are normally used (Lim and Huang, 2007). In spite of having all these efficient adsorbents, the search for abundant and locally available adsorbents with a good oil-sorption capacity is still required for the development of the best available oil-removal technologies.

The local oil-palm industry is estimated to generate 30 million tons of lignocellulosic biomass per year in the form of trunks, fronds, empty fruit bunches and leaves (Sumathi *et al*., 2008). These wastes are not being utilized effectively; land filling and open burning are common practices to eliminate these oil-palm residues, and these methods can cause pollution that adversely impacts the ecosystem (Hashim *et al*., 2010). Therefore, finding uses for these abundant biomasses, especially on a large scale, would be profitable from both an environmental and an economic point of views.

To the best of our knowledge, no work has been conducted on the removal of petroleum hydrocarbons from water by oil palm biomass either for surface or dissolved oil. Therefore, herein we chose oil palm leaves (OPL) as a model adsorbent and studied their potential for removing crude oil from water. Our results are also expected to extend the application of abundant OPL beyond animal feed and fertilizer. However, OPL was reported to consist of 47.7 % holocellulose and 27.4 % lignin by using Wise and Klason method, respectively (Hashim *et al*., 2011). It was also reported that 44.5 % of the holocellulose were α-cellulose. It must be noted that the total percent of the chemical composition is more than 100 % which due to the different method used by previous study. Due to the presence of many hydrophilic groups on the surface of OPL, the surface characteristics of the OPL should be improved to become more hydrophobic for more effective oil adsorption.

Several studies have been conducted for the surface modifications of agricultural waste/byproduct, particularly by using surfactants (Mao *et al*., 2009; Fanta *et al*., 1987), but the use of fatty acids is still rare. Therefore, the present study attempted to use lauric acid (LA) to modify the surface of OPL from polar to hydrophobic and investigate the oil adsorption capacity under various experimental conditions including pH, adsorbent dosage, contact time, initial oil concentration and temperature. The uptake of oil by OPL_{LA} was proved by Fourier-transform infrared spectroscopy (FTIR), field-emission scanning electron microscopy (FESEM) and surface area analysis (BET method). The equilibrium, kinetics and thermodynamics of the adsorption were also investigated.

1.2 Problem Statement

Water pollution by oil has left undesired impact on the environment, aquatic life as well as other living organism. Therefore, an adequate treatment of the polluted water is required. The search for an efficient technology for the removal of oil from aqeuous solution has directed attention to adsorption due to its operational simplicity, low cost requirement and also high effectiveness (Ngah *et al*., 2008).

Due to the high cost and non-biodegrabadility of the current adsorbent, the search for a biodegradable material with excellent adsorption capacity and abundance in local area is still required. Improper handling of oil palm leaf wastes generated from palm oil industry have put an adverse impact on the ecosystem. To the best of our knowledge, no work has been conducted on the removal of petroleum hydrocarbons from water by oil-palm biomass either for surface or dissolved oil. Therefore, in this study we chose oil-palm leaves (OPL) as a model adsorbent and studied their potential for removing crude oil from water.

Hashim *et al*., (2011) reported the characteristics of OPL, which is rich in cellulose, hemicellulose and lignin. It was expected that most of the raw agricultural waste including OPL usually exhibited low oil adsorption capacity due to the low hydrophobicity properties. Thus, it is important to counter this drawback by enhancing the hydrophobicity of the adsorbent. Radetic *et al*., (2008) reported that high oleophilicity is the reason for the outstanding adsorption capacity of most of the currently available commercial adsorbent. Ibrahim *et al*., (2010) also reported the surface modification of raw agricultural waste to boost its performance by increasing the hydrophobicity property of the adsorbent.

Despite the various surface modification methods developed, the role and mechanism of lauric acid in the enhancement of hydrophobicity of OPL have not been discussed yet. Therefore, this study was conducted to investigate the enhancement of crude oil adsorption capacity of OPL by surface modification using lauric acid. In this regard, it is believed that the attachment of lauric acid chain on the OPL will enhance its hydrophobicity property, thus increasing the capability to adsorb more crude oil.

1.3 Objectives of Study

The objectives of this study are :

i. To prepare oil adsorbents from raw OPL.
ii. To modify the hydrophobicity properties of the OPL by using sodium dedocyl sulphate (SDS) and lauric acid (LA).
iii. To study the physicochemical properties of the $OPL_{untreated}$ and OPL_{LA}.
iv. To optimize the removal of crude oil by the OPL_{LA} under various adsorption conditions.
v. To study the equilibrium, kinetics and thermodynamics properties of the crude oil adsorption onto the OPL_{LA}.
vi. To apply the OPL_{LA} to the synthetic crude oil contaminated seawater.

1.4 Scopes of Study

The scope of this study are:

i. Preparation of adsorbent from raw OPL by drying method.
ii. Modification of the hydrophobicity properties of the adsorbent by using sodium dedocyl sulphate (SDS) and lauric acid (LA).
iii. Characterization of the prepared adsorbent by Fourier transform infrared spectroscopy (FTIR), Field emission-scanning electron microscopy (FESEM), surface area analysis (BET) and thermogravimetric analysis (TGA).
iv. Optimization of crude oil removal from aqeuous solution using OPL through batch adsorption system under various parameters including hydrocarbon fraction, surface modification, pH of the oily solution (2-11), adsorbent dosage (0.05 – 1.30 g), contact time (0-60 min), initial crude oil concentration (0-6400 mg L^{-1}) and temperature (313-323 K).
v. Investigation of adsorption equilibrium isotherms using the Langmuir, the Freundlich and the Temkin equilibrium models. The kinetics data was subjected to kinetic study using Lagergren-pseudo-first-order and Ho-psuedo-second-order equations and intraparticle diffusion model. The thermodynamic parameters, which are the changes in enthalpy (ΔH^o), entropy (ΔS^o) and standard free Gibbs energy (ΔG^o) were calculated based on the van't Hoff equation. Lastly, the Arrhenius equation was used to calculate the activation energy of the adsorption.
vi. Application of the system to synthetic crude oil contaminated seawater using seawater obtained from Teluk Ramunia.

1.5 Significance of Study

This work was conducted to study the adsorption of crude oil onto OPL. Utilization of OPL, a waste from palm oil plantation for potential oil adsorbent reduced the overall preparation costs. In addition, this effort also helped in the elimenation of this waste in a proper way, thus minimizing environmental pollution. On the other hand, the effective removal of oil spills from the seawater is a problem of great importance and interest for the society worldwide. Due to the modification of the adsorbent, OPL demonstrated high ability of adsorbing crude oil from aqueous solution. In terms of efficiency and effectiveness, this oil sorbent rapidly removed crude oil from crude oil contaminated seawater. As a consequence, modified OPL was proven to boost a great and potential oil adsorbent for oil spill clean-ups, as well as its contribution to the green and sustainable technology.

1.6 Thesis Outline

This thesis was divided into five main chapters. In Chapter 1, the introduction for the overall study was discussed. Problem statement of the current research was stated to stress on the reason why this research must be carried out. Objectives and scopes of the study were clearly stated in the section 1.3 and 1.4, respectively. In section 1.5, significance of the study was stated.

In Chapter 2, all the literature reviews related to the current phenomenon of oil spill, and treatment technologies of oil spill removal were discussed. In addition, this chapter also concluded several basic principals related to adsorption process and their equilibrium, kinetics and thermodynamics studies. Extensive surveys on previous study related to adsorption of oil by various types of adsorbents were also discussed in this chapter.

Chapter 3 described all the methodologies related to current research. Detailed information on the materials and chemicals used in this study were also listed. This chapter also included the detailed methodologis of the preparation, modification and also the adsorption of crude oil onto OPL.

Chapter 4 discussed all of the experimental results obtained from the preliminary study comprehensively. The first part of this chapter involved the characterization of OPL followed by the adsorption of oil under various parameters. Finally, the equilibrium, kinetics and thermodynamics studies were discussed.

Finally, in Chapter 5 the conclusions obtained from this research were reported. Some recommendations for future works were also included in this chapter.

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